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ADVANCED EXCITATION TECHNIQUES FOR TUNABLE INFRARED LASERS

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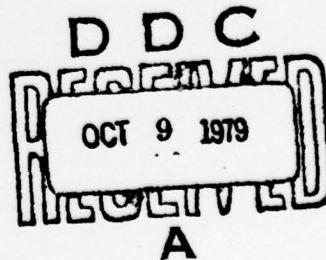
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Raman mixing, tunable infrared laser, optical parametric oscillator, hydrogen and methane Raman medium

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
This report describes the theoretical concept and experimental progress initiated to devise a tunable laser source, broadly tunable over the middle infrared wavelength range. The concept derives from frequency conversion by scattering from coherently driven Raman vibrations in hydrogen or methane gas. This investigation concentrated on the feasibility of producing a wide-band tunable, infrared source using a designator energy →next page

category Nd:YAG pump laser to simultaneously drive a lithium niobate parametric oscillator and coherent Raman mixing in gaseous media.

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ADVANCED EXCITATION TECHNIQUES FOR TUNABLE
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PHASE I - CONCEPT AND FEASIBILITY EVALUATION

1. INTRODUCTION AND BACKGROUND

This report describes the theoretical concept and experi-
mental progress initiated to devise a tunable laser source,
broadly tunable over the middle infrared wavelength range.

The mid-infrared atmospheric transmission band extends from
8 to 20 micron, and has proven so far to be the most significant
atmospheric window for military operations apart from the visible
band. The highly efficient carbon dioxide laser is operable on
various, discrete transitions at 9.5 to 11.5 microns. However,
existing data on optical transmission of battlefield smoke and
aerosol obscurants strongly indicate the need for a broadly
tunable laser source operable within the total limits of the
atmospheric window. This program supports this requirement, and
attempts to prove the concept and feasibility of designing a
continuously tunable laser source which will allow experimental
observation of absorption and scattering from battlefield atmos-
pheric polutants within this significant spectral region.

Giordmaine and Kaiser[1] first reported the process of
frequency conversion by scattering from coherently driven Raman
vibrations in calcite. This coherent Raman mixing process involves
the interaction of four radiation fields in an appropriate non-
linear medium. Two of these fields are externally provided; a
"strong," fixed frequency pump, and a "weak," tunable pump.
The role of the "strong" laser pump field is to initially produce
stimulated Raman scattering in the medium and thus provide a
local oscillator at the Raman frequency for (subtractive) mixing
and frequency conversion of the "weak" tunable source. The
frequency conversion process is dictated by the energy conserva-
tion equation

$$w_t - w_t' = w_p - w_p' \tag{1}$$

where w_t is the tunable input frequency, w_t' is the output
frequency, w_p the "strong" pump frequency, and w_p' is its Stokes
shifted, Raman scattered frequency.

1. J. A. Giordmaine and W. Kaiser, Phys. Rev., Vol. 144,
p 676, 1966.

Since $w_p - w_p' = w_R$, the Raman frequency (which is simply the characteristic vibrational frequency of the medium), the equation reduces to

$$w_t' = w_t - w_R . \tag{2}$$

That is, under the initial Raman scattering influence of the "strong," fixed-frequency pump, the net process appears as if the "weak," tunable pump is also separately Stokes shifted by the Raman frequency.

The advantage of the CRM (coherent Raman mixing) process lies in the relative intensities of the pump fields necessary for tunable output generation. In essence, the only role of the "strong" pump is to exceed SRS (stimulated Raman scattering) threshold in the medium. As a result, the "weak" pump power threshold to produce its Stokes component can be lowered by several orders of magnitude.[2] Thus, any convenient "strong" pump source can be utilized at a fixed-frequency, without the need to generate strong tunable radiation at a power level sufficient to alone produce SRS. The CRM process therefore represents a powerful generic technique for production of tunable radiation. G. V. Venkin, et al[2] showed that 50% energy conversion could be achieved with fixed-frequency biharmonic pumping of hydrogen with a pulsed dye laser and its second-harmonic. S. G. Brosnan, et al[3] extended this concept using a neodymium laser and parametric oscillator, with hydrogen, to generate continuously tunable infrared generation.

A qualitative and semi-quantitative understanding of the CRM process may be gained by a description of the physical model adopted for Raman scattering, rather than an attempt to present the rather formidable mathematical formulation that follows from it. Raman scattering, like the familiar Rayleigh scattering process, involves the re-radiation of incident radiation by the oscillating dipoles formed by the positively charged nuclei and surrounding electronic charges that compose the (transparent) medium. With Rayleigh scattering, the incident field excites oscillating dipoles which coherently reradiate the same frequency. The polarization or dipole concentration

2. G. V. Venkin, G. M. Krochik, L. L. Kulyuk, D. I. Maleev, and Yu. G. Khronopulo, Sov. Phys. JETP, Vol. 43, p 873, May 1976.

3. S. J. Brosnan, R. N. Fleming, R. L. Herbst, and R. L. Byer, Appl. Phys. Lett., Vol. 30, p 330, 1 April 1977.

4. G. Placzek, "Marx Handbuch der Radiologic," E. Marx, Editor, 2nd edition, Vol. 6, Leipzig Acadamiche Verlagsgescell-schaft, 1935, part II, p. 205.

is linearly proportional to the incident field amplitude by the electronic polarizability. If, however, the nuclei are undergoing some periodic motion as in the characteristic vibrational modes of a molecule, the electronic polarizability is modulated by the same period and the motion of the radiating dipoles is modified by the relative position of the nuclei.[4]

This reradiation or scattering process differs from the laser process in that there is no absorption of an incident photon and excitation of the medium to some characteristic electronic state. In terms of energy levels, the excited initial state is viewed as a virtual energy level, equal to the energy of the incident photon, as shown in Figure 1.

The incident pump radiation, at frequency w_p, excites the system to a virtual level, which then decays to a terminal state corresponding to a vibrational level of the medium. The radiative decay corresponds to emission of a Stokes-shifted photon of lower frequency w'_p. The system then decays in phonon emission at frequency W_R, characteristic of the medium. This emission at W_R provides the source for mixing with a second input frequency, the tunable input at W_T, to produce the tunable, Stokes-shifted output.

2. DESIGN CONSIDERATIONS

The experimental design which chooses pump laser frequency and nonlinear medium is dictated by the requirement for the tunable output frequency. In addition, design considerations for the investigation reported here were restricted by the availability of necessary components and abbreviated project duration. Due to these restrictions, major emphasis was placed on an investigation of the generic technique of CRM to at least provide a technoligical basis for possible future undertaking of this project.

In order to enhance compatibility of this investigation with practical application, the pump laser was chosen to be of the target designator energy category.

Based on these considerations, the experimental system used a Q-switched Nd:YAG laser as the "strong" pump and high-pressure hydrogen or methane was selected for the nonlinear medium. Besides being readily available and relatively cheap, the choice of a gaseous nonlinear medium has advantages over solid-state devices. Because gases have low dispersion over broad spectral ranges, phase matching problems are alleviated. With the high power pump utilized, the gain is high enough so that conversion efficiency is attained within a distance short compared to the coherence length produced by phase mismatch.[3] Secondly, internal radiation damage is not a problem as it is with solid or liquid media. Although some liquid media possess attractive characteristics for SRS due to their low scattering thresholds, they are typically highly toxic and undesirable from that aspect.

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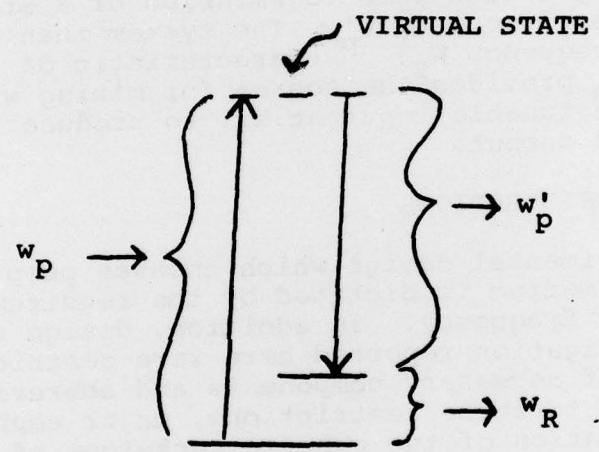
Figure 1.   Energy level concept of Raman scattering.

The major experimental difficulties working with gaseous media arises from the high pressures involved (as much as 100 atmospheres) and their flammability - hydrogen may self ignite in a high pressure leak.

The experimental setup is shown in Figure 2. The Nd:YAG laser emits 90-100 mJ Q-switched pulses, of approximately 12 ns FWHM duration, at 1.064 μm. Output is multi-transverse mode in a 1/4" diameter near-field beam. This radiation is used first to excite a lithium niobate, angle-tuned OPO (optical parametric oscillator), whose output is tunable, either in the signal or idler wave, from 1.8 to 2.6 μm. At the output of the OPO, approximately 1.5 mJ of this tunable radiation, together with the transmitted 1.064 μm beam is used as the strong pump radiation, and the tunable component as the weak pump. These two beams are focussed into a 30 cm long, stainless steel gas cell, through a 1/4" thick quartz window used at the input port. The gas cell windows were uncoated and no external mirrors were employed, so that the SRS system was in a single-pass configuration. Table I shows the extent of continuously tunable wavelengths possible with this choice of pump input and hydrogen or methane gas. The output wavelength is derived from Eq. (2).

Table 1. CRM output wavelengths vs OPO input.

	OPO (μm)	Hydrogen (4.55 cm^{-1})	Methane (2914 cm^{-1})
Signal	1.8	7.14	3.79
Degeneracy	2.13	18.52	5.62
Idler	(2.4	---	10.77
	(2.6	---	10.77

Note that the CRM output wavelengths tend to very large values as the OPO frequency approaches the Raman frequency for the medium.

3. EXPERIMENTATION

a. Stimulated Raman Scattering

Before an attempt was made to attain CRM, numerous preliminary experiments were carried out to separately achieve SRS with hydrogen or methane in a single-pass system, with the available pump energy.
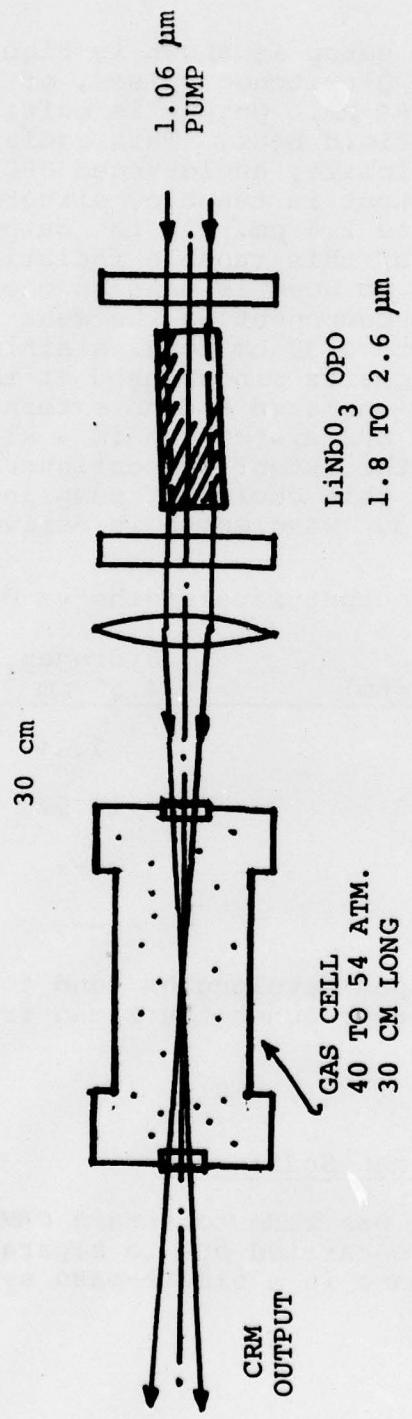
Figure 2. Schematic diagram of coherent Raman mixing experimental setup.

SRS was first achieved with hydrogen at pressures greater than about 32 atm (470 PSI). The strong I-Stokes component was detected at 1.91 μm, and I and II - Anti-Stokes components at 0.738 μm and 0.565 μm were visually observed. SRS could not be attained at lower pressures even with longer gas cells and variation of lens focal length. This fact produced an unfortunate circumstance because neither the salt or germanium windows available which could transmit long wavelength radiation were strong enough to withstand the high gas pressures. Consequently, methane was substituted as the SRS medium, and a 2 mm thick sapphire window was used in the output port. Sapphire transmits to 6 μm, which is appropriate for the tuning range of the OPO.

Methane introduced new problems because of its higher SRS threshold. SRS threshold was achieved at pressures greater than about 44 atm (650 PSI). Due to the lack of a high pressure gas regulator, operating pressure in the gas cell was obtained through a leak valve directly from the methane cylinder. Attempts to increase gain by increasing methane pressure were experimentally limited to about 54 atm (800 PSI).

An interesting phenomenon was observed with both methane and hydrogen in these preliminary SRS experiments. The initial setup used a 20 cm focal length lens to focus the pump radiation into the gas cell, and the pump laser was operated at 10 PPS repetition rate. With hydrogen, only about 50% of the input pulses achieved SRS. When methane was used, this reliability rate dropped to only about 10%. This factor remained about the same regardless of the gas pressure. Increasing the lens focal length improved the situation, but it was found that the pump rep-rate had the greatest effect - 80% to 90% reliability was achieved only when the rep-rate was reduced to 2 PPS. Best SRS output occurred with a 30 cm focal length lens. Thus, for the given pump power input, an optimum pump intensity and rep-rate is dictated. The rep-rate dependence is apparently caused by the low thermal conductivity of the gaseous media, with consequent thermal blooming and optical turbulence within the region of the focussed pump beam.[5,6] Methane was more sensitive in this regard than hydrogen, reflecting their difference in thermal conductivity. In addition, it was discovered that SRS output seemed to be sensitive to slight adjustment of the pump laser (plane) cavity mirrors, even though their adjustment showed no observable difference in energy output of the pump beam on a separate monitor. This effect has been observed by Meadors[7]

5. C. D. Decker and P. J. Gardner, "Efficient Raman/Dye Laser Radiation Source," Technical Report AFAL-TR-78-58, May 1978, Wright Patterson AFB.

6. J. G. Meadors and M. A. Poirier, IEEE J. Quant. Elect., Vol. QE-8, p 427, April 1972.

7. J. J. Meadors and M. A. Poirier, "Stimulated Raman Scattering in Methane with Nd:YAG Pump," Final Technical Report 4297-1, Contract DAAG29-76-G-0140, June 1977, US Army Research Office.

and ascribed to transverse mode dependence on cavity mirror alignment. Perhaps small changes in mirror alignment produce a more Gaussian-like beam profile, that induces self-focussing of the pump beam, with increased intensity and SRS gain. In the absence of optical turbulence, it is estimated that the pump beam is focussed to a spot of about 1 mm diameter, producing over 100 MW/cm^2, well over SRS threshold for either gaseous medium.

A fast, S-1 response, vacuum photodiode and Tektronics 7904 oscilloscope were used to monitor the transmitted pump pulse during SRS operation in an effort to observe pump beam depletion. Although depletion was observed, it was too irregular from shot-to-shot to be useful for any quantitative determination. For qualitative purposes, it appeared that the SRS occurred approximately symmetrical about the peak of the pump pulse. Its position is affected by thermal loading and competitive effects, such as stimulated Brillouin Scattering, which occur at these operating pressures.[5-7] The relative position of the SRS pulse, with respect to the OPO output, is important for efficient CRM operation. Ideally, they should be produced simultaneously in order to attain complete overlap. However, OPO operation was found to occur mostly on the trailing edge of the pump pulse (due to finite radiation buildup time in the OPO cavity) and thus is mistimed, to some degree, with respect to the SRS pulse.

b. Optical Parametric Oscillator

An angle-tuned, OPO (optical parametric oscillator) was set up in the configuration shown in Figure 2. The OPO[8] was a 5 cm long lithium niobate crystal with a 7 cm spaced cavity consisting of two plane mirrors with high reflectivity centered at the degenerate wavelength of 2.13 μm. Each mirror had about 90% transmission at the 1.06 μm pump wavelength. The measured OPO output was at least 1.5 mJ, with 5 to 7 ns FWHM pulse duration, giving approximately 250 kW peak power, over a tunable range of 1.8 to 2.6 μm. Observation of the transmitted pump beam showed that depletion, hence OPO pulse duration, occurred mostly on the trailing edge of the pump pulse waveform.

c. Coherent Raman Mixing

The OPO, focussing lens, and methane gas cell were set up along the axis of the pump beam as shown in Figure 2. An available (hydrogen fluoride) laser spectrum analyzer was modified for use with the CRM experiment. It was essentially a 0.5 m Ebert mount diffraction grating spectrograph, originally using a wavelength scale and a quenched fluorescent screen for wavelength observations. The fluorescent screen was removed and replaced with external detectors to separately monitor OPO wavelength and expected CRM output wavelength calculated from Eq. (2). An indium arsenide detector was used for the OPO

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8. T. F. Ewanizky, IEEE J. Quant. Elect., Vol. QE-14, p 962, Dec. 1978.

and a pyroelectric energy detector for the CRM wavelength. CRM output, expected at wavelengths up to 6 μm, was marginally detectable on the lowest (2 μJ) scale of the pyroelectric detector readout, because of inherent transient noise and the low reliability of the necessary SRS process. Low relative energy output was expected for the CRM process, but peak power could be reasonably high, perhaps in the kW range. No high-speed detector with appropriate spectral bandwidth was available to display output power rather than energy.

4. ANALYSIS OF RESULTS

   a. The output power and beam quality of the pump laser was marginal for the experiments. Together with uncoated optics and high-pressure requirements on the output window, definitive measurement and analysis were severely limited.

   b. The relative timing and short overlap of the SRS and OPO durations must definitely be regarded as a prime source for low efficiency.

   c. The relatively high pressures required in both hydrogen and methane for SRS excitation by the 1.06 μm pump radiation produces deleterious effects even at relatively low rep. rate.

5. CONCLUSIONS AND RECOMMENDATIONS

   All of the points covered in Part 4 could, in principle, be taken into account for a second effort. It would appear that the degrading factors discovered can be ameliorated and still retain the beneficial aspects of CRM concept as a means to generate continuously tunable infrared radiation at high peak powers. Based on the experimental results, the necessary improvements can be enumerated as follows:

   a. Increase pump power and beam quality, and optimize cell and OPO optics. This will allow a reduced OPO threshold and operation over a longer portion of the pump pulse. In addition, the pump pulse could be separated by a beam splitter placed before the OPO and recombined after it as an optical delay to improve the OPO/SRS timing overlap.

   b. Down collimate the input radiation rather than focussing could increase the gain length for SRS.

   c. Assuming parts a and b were accomplished, lower gas pressure in the cell would be required for SRS threshold. This factor would allow an increase in rep. rate, especially so if hydrogen were used as the nonlinear medium.

   d. Hydrogen would appear to be preferable to methane for CRM because of lower SRS threshold and greater insensitivity to thermal/optical turbulence.

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e. The CRM concept should be extended, perhaps with a pulsed carbon dioxide laser as the strong pump source and Raman scattering from the hydrogen rotational levels.
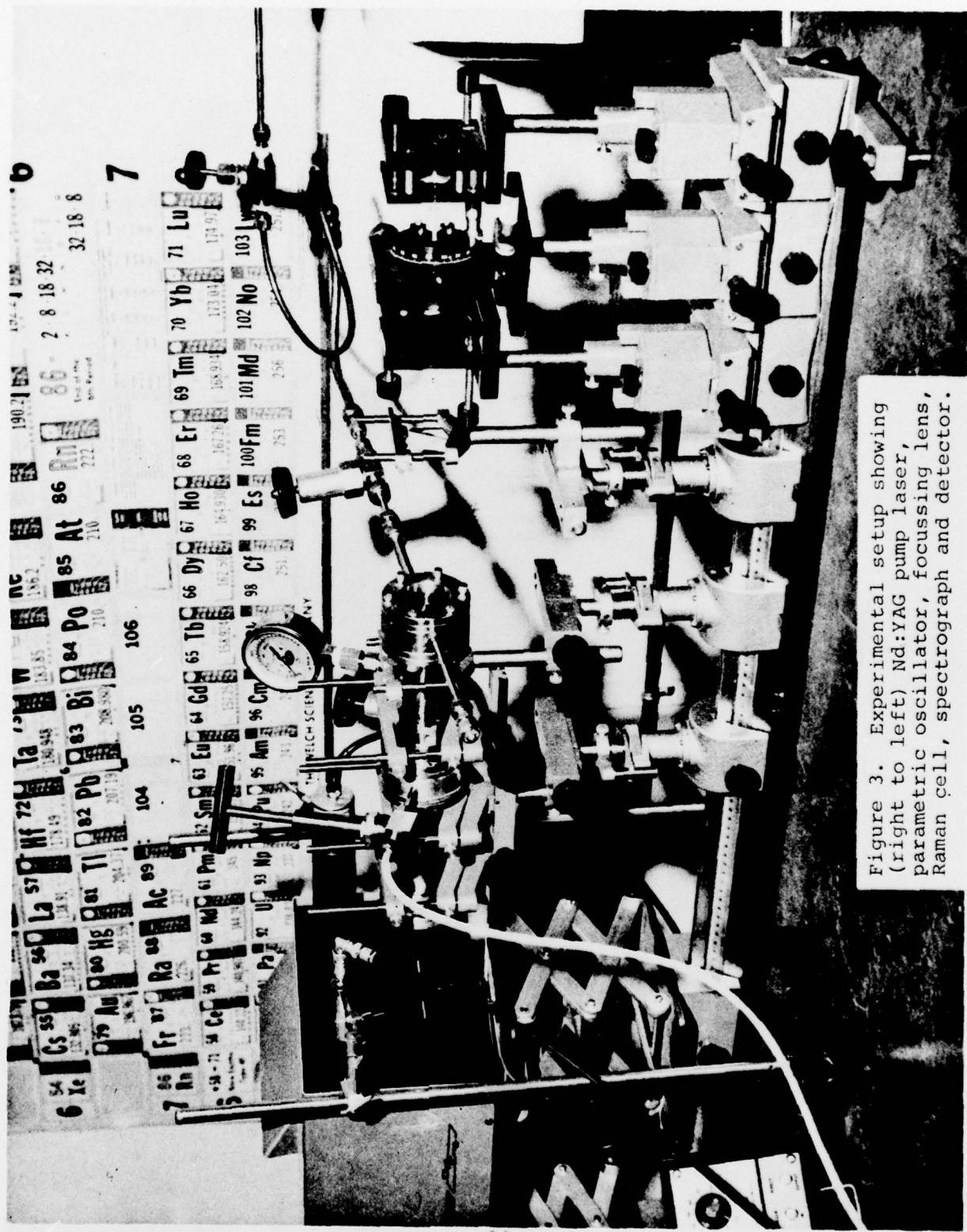
Figure 3. Experimental setup showing (right to left) Nd:YAG pump laser, parametric oscillator, focussing lens, Raman cell, spectrograph and detector.

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